Supporting Information for

"Pulsed Growth of Vertically Aligned Nanotube Arrays with Variable Density"
by Jeremy J. Jackson, Alex A. Puretzky, Karren More, Christopher M. Rouleau, Gyula Eres, and David B. Geohegan

1. Illustration of the Approximate Fitting Procedure to Evaluate Optical Extinction Coefficients

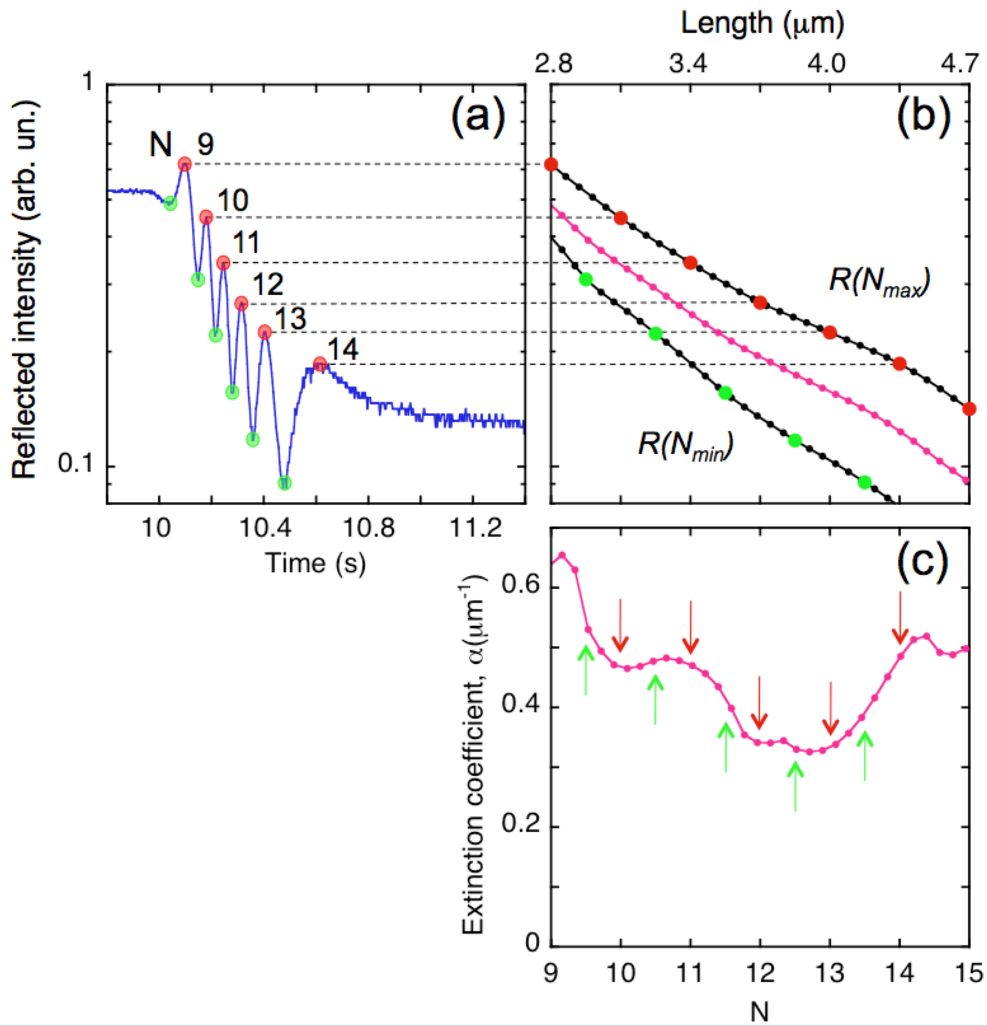

Figure S1. Illustration of the approximate procedure for evaluation of effective extinction coefficients. (a) An example of time resolved reflectivity oscillations measured *in situ* during the second gas pulse (see Fig. 2(b), curve 2) showing the interference maxima from $N = 9$ to 14. (b) Reflected intensities measured at the maxima, $R(N_{max})$, and minima $R(N_{min})$ are plotted versus the number of oscillations, N (large points). Additional (small) points, obtained as a result of linear interpolation of the $R(N_{max})$, and $R(N_{min})$ are used to calculate the average reflectivity curve, $R(N)$, which is shown in the middle. (c) An effective extinction coefficient obtained as the slope of the averaged curve, *i.e.,*

$\alpha = -\dfrac{d\left[\ln(R/R_0)\right]}{d(2d_0N)}$. The arrows show the positions of the measured maxima (upper arrows) and minima (lower arrows).

2. Comparison of Density Evolution in VANTAs : Continuous vs. Variable Feedstock Flux

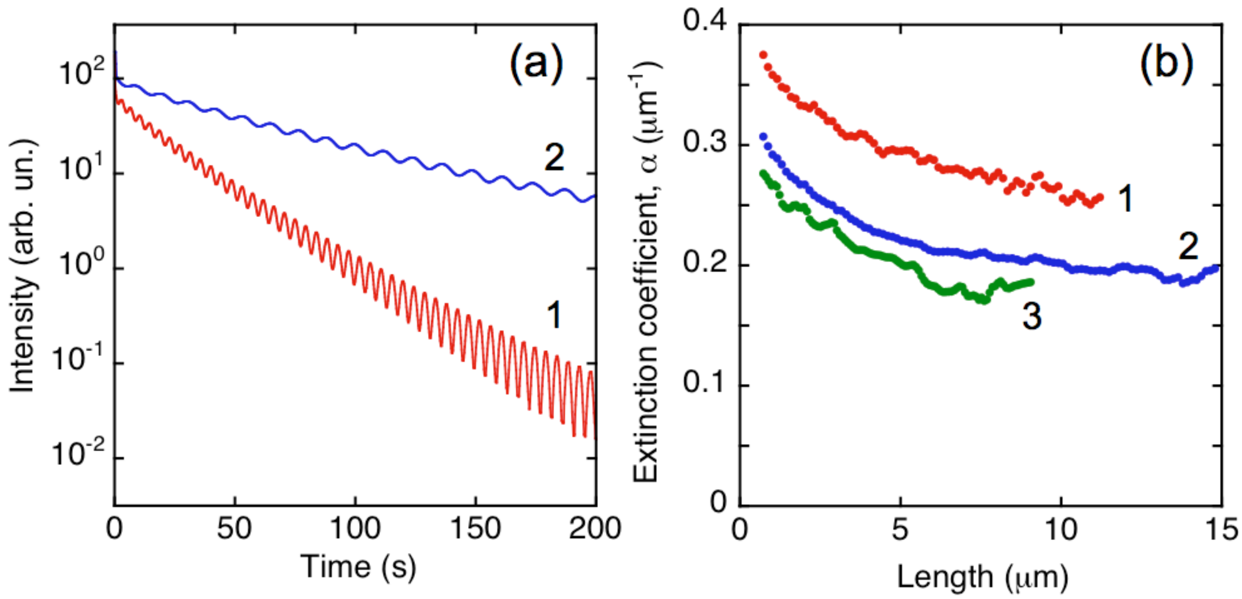

Figure S2. (a) Time resolved reflectivity curves measured during VANTA growth in a low pressure CVD reactor at 720 °C using 10 nm Al_2O_3 and 0.5 nm Fe films deposited on a Si substrate. The total pressure was 6.3 Torr with the partial flow rates of C_2H_2:10 sccm (curve 1), 5 sccm (curve 2), H_2: 250 sccm, and Ar: 2000 sccm. **(b)** Comparison of the effective extinction coefficients measured *in situ* for the two reflectivity curves shown in (a) with the case of variable acetylene flow (curve 3), when C_2H_2 flow was switched from 10 to 5; 5 to 2; and 2 to 1 sccm, respectively (see Figs. 4(b) and 5(c)).

Figure S2(a) shows representative time-resolved reflectivity curves measured for two different continuous growth experiments of VANTAs from argon/hydrogen/acetylene mixtures with constant acetylene supplies of 10 sccm (red curve 1) and 5 sccm (blue curve 2). Each fringe on the curves corresponds to 300 nm growth of the nanotube array. These can be compared with the data of Figure 4 which shows the reflectivity data for an array where the acetylene flux was changed three times within a single run to study growth rates and extinction coefficient evolution at acetylene partial flows of 10 sccm, 5 sccm, 2 sccm, and 1 sccm for the same catalyst.

Figure S2(b) compares the declines in the optical extinction coefficient derived from the two constant-flux curves of Fig. S2(a) with the decline for the multiple-flux data of Fig. 5(c). Although each array grows at a different rate (or at multiple rates as shown in Fig. 4), the optical extinction coefficient for the layers grown later in each run (by base growth at the substrate) displays a similar decline with length, *d(t)*. The absolute magnitude of the optical extinction coefficient in each case is different, as might be expected from differences in VANTA density arising from catalyst preparation from run to run.